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Title: **APPLICATION OF THE SOURCES CODE IN NUCLEAR SAFEGUARDS**

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APPLICATION OF THE SOURCES CODE IN NUCLEAR SAFEGUARDS

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ABSTRACT

The Sources Code System provides a greatly expanded calculational capacity in the field of nuclear safeguards. It is becoming more common that we are called upon to perform assays on materials for which no standards exist. These materials tend to be mixtures of nuclear materials and low-Z compounds (spent fuels in a variety of matrices, in-process compounds such as UF_6, MOX with varying water content). We will present some applications of the Sources Code and discuss the application calculated (α,n) source terms in neutron coincidence counting for nuclear safeguards

INTRODUCTION

Nondestructive assay (NDA) of nuclear material using passive neutron counting is a very common technique. By employing neutron coincidence counting and multiplicity counting, sufficient observables can be obtained to accurately determine the mass of nuclear material present in a given container. Oxides and fluoride compounds have long been a problem in NDA measurements because of the bias introduced by an unknown contribution to the neutron source term, both as a direct contribution to neutron singles events and by way of sample multiplication in doubles and triples events.

We will present the "Point Model" Equations, derived by Böhnel[1] and modified by Krick and Enniselin[2], that are the technical backbone of passive neutron counting for NDA application. We will then present a series of real world applications where the Sources Code has been used to calculate the (α,n) contribution to particular source terms

SAFEGUARDS NEED FOR (α,n) SOURCE TERMS

In typical safeguards applications there are two means of producing a neutron; fission and (α,n) reactions. Neutrons are ideal for NDA measurements because they readily penetrate through most containers and matrix materials. When a material fissions; one two or three neutron are produced in accordance with well known probability distributions for each fissioning species. The mass of an actinide material can be determined from the neutron production rate of the sample. The Point Model equations describe this process by predicting the rate of singles (S), doubles(D), and triples(T) neutron production are provided below.

$$S = mF\varepsilon M\upsilon_{s1}(1+\alpha) \tag{1}$$

$$D = \frac{mF\varepsilon^2 f_d M^2}{2}\left[\upsilon_{s2} + \left(\frac{M-1}{\upsilon_{i1}-1}\right)\upsilon_{s1}(1+\alpha)\upsilon_{i2}\right] \tag{2}$$

$$T = \frac{mF\varepsilon^3 f_t M^3}{3!}\left\{\begin{array}{l}\upsilon_{s3} \\ +\left(\frac{M-1}{\upsilon_{i1}-1}\right)\left[3\upsilon_{s2}\upsilon_{i2} + \upsilon_{s1}(1+\alpha)\upsilon_{i3}\right] \\ +3\left(\frac{M-1}{\upsilon_{i1}-1}\right)\upsilon_{s1}(1+\alpha)\upsilon_{i2}^2\end{array}\right\} \tag{3}$$

Where,
S = singles neutron counting rate (n/s)
m = mass of nuclear material (g)
F = specific spontaneous fission rate (n/s/g)
ε = detection efficiency
M = sample leakage multiplication
α = ratio of (α,n) to spontaneous fission neutron production
f_d = fraction of fission neutrons arriving in the doubles coincidence gate
f_t = fraction of fission events arriving in the triples coincidence gate

υ_{s1}, υ_{s2}, υ_{s3} = first, second, and third reduced moments of the spontaneous fission multiplicity distribution

υ_{i1}, υ_{i2}, υ_{i3} = first, second, and third reduced moments of the induced fission multiplicity distribution

In each of the equations 1, 2, and 3 the term, α, appears. As defined, α is the ratio of (α,n) to spontaneous fission production in a sample matrix. It is this term that we obtain from the Sources program. In theory, because we have three equations, we could measure an unknown sample and determine m, M, and α without *a priori* knowledge of the sample or the matrix. In practice, in order to accurately measure, T, either a very high detector efficiency or a very long count period is required. Typically, we settle for observing S, and D. This requires *a priori* knowledge of either M or α to determine the material mass.

We have built numerous systems where we have employed the so-called "Known-α" solution to the series of equations 1 and 2 for m and M. Some of those applications have required the application of the Sources code to determine the value of α because of situations where no physical standard could be made or that the calibration sample production costs would be prohibitive. In those cases we have calibrated our NDA systems using the Sources program and Monte Carlo codes.

APPLICATIONS

A. UF_6 In Enrichment Facilities

Uranium has a very low spontaneous fission rate and is generally not assayed using passive neutron measurement. However, in certain chemical forms the (α,n) production becomes large enough that passive neutron measurement is a practical approach. One of these chemical forms is UF_6.

NDA technicians have long tried to use passive neutron counting techniques to measure in-process material holdup in uranium enrichment cascades. These attempts have met with limited success because users have applied traditional holdup measurement approaches and attempted to resolve individual components and sum the results to determine the material inventory in an entire hall.

^{238}U produces the vast majority of the spontaneous fission neutrons from uranium (13.6 n/s-kg), a number too small to permit the accurate passive neutron assay of small quantities of uranium. However, in the case of UF_6, a significant (α,n) neutron production from $^{234}U(\alpha,n)$ reactions on the fluorine (5800 n/s-kg). If we balance these yields by the naturally occurring isotopics of uranium the neutron yield of UF_6 becomes 45.7 n/s-kg U as compared to the 13.7 n/s-kg U for metallic uranium. In the case of a uranium enrichment hall where tons of UF_6 are typically present, the specific activity of UF_6 is adequate to allow NDA measurement of an entire hall.

With an accurate specific activity for UF_6 feed material and energy spectrum calculated from the Sources Code, shown in Figure 1, we then set out to calibrate the response of our detector system in a large hall containing a generally homogeneous distribution of UF_6 using Monte Carlo Neutron Transport codes. The determination of the neutron energy spectrum from the Sources Code is essential to obtaining good results from the Monte Carlo simulation. Once calibrated, we then sample the neutron continuum in the hall with our detector system and determine the total uranium holdup in the entire hall in a fraction of the time required for traditional holdup measurement techniques. We have applied this method in two enrichment facilities over multiple years with excellent results and resulting in significant time savings for both the operators and the international inspectorate.

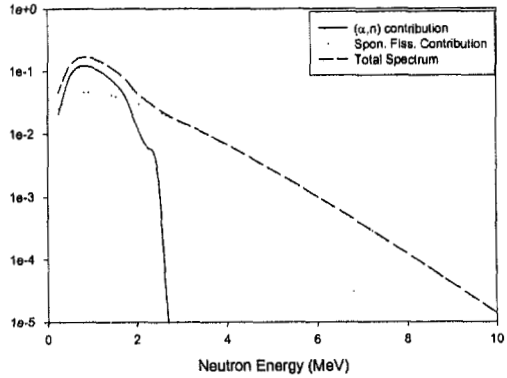

Figure 1. Neutron energy spectrum in natural UF_6.

B. Moisture content in MOX

Mixed oxide fuels (containing both Pu and U oxides) are used in many nuclear fuel cycles around the world. The value of α in these materials have been measured in multiplicity counters for small samples (counters able to measure T, and hence determine α). Those results have been extrapolated to application to in-facility measurements of bulk MOX material with reasonable success. With the application of the Sources Code, these precarious extrapolations are no longer necessary. The difficulty of using a Known-α measurement on MOX materials has come to light recently when we examined the effect of an unknown moisture content in the MOX powder on the neutron production rate. Obviously, as the moisture content of the MOX increases the (α,n) production increases. The value of α in Eqs. 1,2 and 3, determined from dry laboratory samples, introduces a bias in the resulting assay mass.

To resolve the assay bias issue, we used the Sources Code to determine the value of α as a function of moisture content in MOX. The results of that calculation are presented in Figure 2. Of equal utility, the spectral neutron energy data provided by the Sources Code for each calculated moisture level was used to optimize the detector design to provide a means of determining the moisture content of the MOX and thus, providing the correct value of α for use in Eq 2 to determine the mass of material.

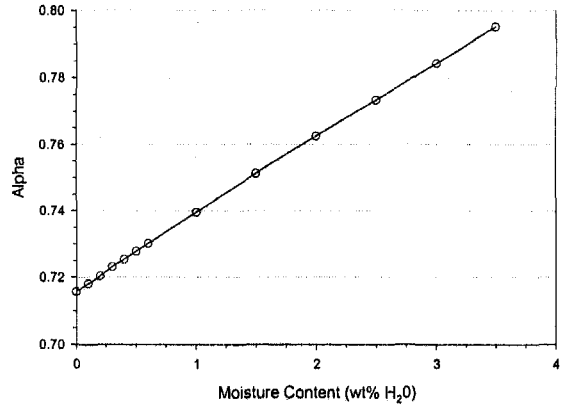

Figure 2. Trend in α with moisture content in high burnup MOX

As the moisture content of MOX powder increases, the neutron energy spectrum is shifted to higher energy. This effect is shown from our Sources Code calculations in Fig. 3. The shift in the neutron energy spectrum has allowed us to optimize the tube placement in the detector to monitor the energy spectrum to determine the moisture content and thus a corrected value of α on a sample-by-sample basis.

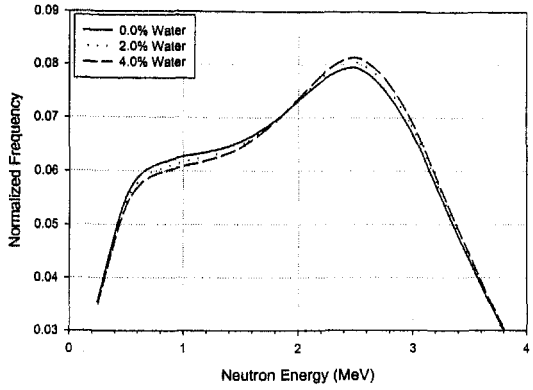

Figure 3. Neutron spectral shift with moisture in MOX.

C. Vitrified Waste

As a third example of our applications of the sources code we consider the safeguarding of vitrified nuclear waste. In this case the facility operator declares the quantity of plutonium contained in a container of vitrified nuclear waste. The operator would like to get credit for the disposal of the declared mass of plutonium. Because the vitrifying matrix is basically borosilicate glass, there is a diversion scenario that the operator could add less plutonium than the declaration and increase the boron fraction in the glass to produce a neutron source term that agrees with the declaration.

For this project we used the sources code to examine the neutron spectral shift caused by the proposed diversion scenario. Because the actinide signal is dominated by ^{244}Cm for this waste, we used the ^{244}Cm signal in conjunction with a verified Cm/Pu ratio to determine the Pu content of the waste container.

Figure 4 shows the neutron spectrum for a typical waste container having 11 weight percent boron. If the operator attempted to divert material by the proposed scenario the high energy peak from the (α,n) reaction on boron increases dramatically to offset the reduced ^{244}Cm signal.

We then installed a specialized detector system that would be sensitive to the calculated energy shift to prevent this diversion.

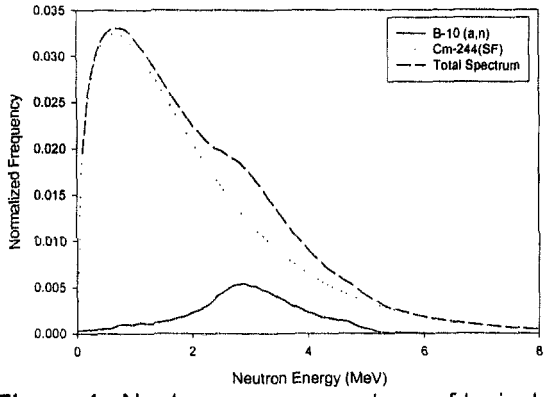

Figure 4. Neutron energy spectrum of typical vitrified waste (11% boron).

CONCLUSIONS

The Sources Code System is a singularly useful tool for safeguards applications. As we are called upon to develop NDA equipment for increasingly challenging chemical forms and matrices, the application of this program for physics assessment of new detection problems will only increase.

Over the years we have employed this code, the evolution from pet project to a truly useful design tool has been remarkable. We no longer are reliant upon empirical trends or assuming similarity to measured cases to accommodate the α term in the basic equations of passive (and active) neutron coincidence counting.

REFERENCES

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2. M.S. Krick, "^{240}Pu-effective Mass Formula for Coincidence Counting of Plutonium with Shift Register Electronics," in "Nuclear Safeguards Program Status Report, May-August 1977," J.L. Sapir, Comp., Los Alamos Scientific Laboratory Report LA-7030-PR (1977), p.16.